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A MOBILE MELT-DILUTE MODULE FOR THE TREATMENT OF ALUMINUM RESEARCH REACTOR SPENT FUEL

Harold Peacock, Donald Fisher, Thad Adams, Robert Sindelar, and Natraj Iyer
Westinghouse Savannah River Company
Savannah River National Laboratory
Aiken, South Carolina, USA

and

David Sell, Ken Allen, Eric Howden, and Brian Westphal
Argonne National Laboratory, West
Engineering Technology Division and Nuclear Technology Division
Idaho Falls, Idaho, USA

ABSTRACT

A mobile melt-dilute (MMD) module for the treatment of aluminum research reactor spent fuel is being developed jointly by the Savannah River National Laboratory and Argonne National Laboratory. The process utilizes a closed system approach to retain fission products/gases inside a sealed canister after treatment. The MMD process melts and dilutes spent fuel with depleted uranium to obtain an isotopic content of less than 20%. The final ingot is solidified inside the sealed canister and can be stored safely either wet or dry until final disposition or reprocessing. The MMD module can be staged at or near the research reactor fuel storage sites to facilitate the melt-dilute treatment of the spent fuel into a stable non-proliferable form.

1. Introduction

The recent international focus on nuclear non-proliferation includes consideration of the spent fuel located in eighteen countries with Russian designed research reactors located outside the border of Russia. Highly-enriched uranium (HEU) fuel assemblies have been used to operate these reactors, and the spent fuel assemblies are stored in cooling ponds. These HEU assemblies pose a weapons proliferation concern. In addition, these research reactors and spent fuel storage sites typically do not have the high level of security infrastructure appropriate for protecting such fuel. While the U.S. has taken steps to help improve the security infrastructure at these sites, it would be preferable to ensure that the HEU fuel is in a non-proliferable form.

The Mobile Melt-Dilute (MMD) treatment process is an attractive option that can treat spent fuel by converting the weapons useable HEU to a safe and secure low-enriched uranium (LEU) ingot through dilution with depleted uranium. The process is modular, reusable, and readily portable to a desired site or storage location.

The basic Melt-Dilute (MD) technology was developed to treat foreign and domestic spent research reactor fuels returned to the Savannah River National Laboratory in the1990’s. This same technology is adapted for the mobile process except that the spent fuel is processed in a closed canister so fission products along with the low enriched uranium (LEU) ingot are retained inside a sealed canister[1-2]. Proof of principle experiments and component evaluations are being performed as part of an integrated approach for flowsheet development and design and fabrication of prototype system components.

2. Background

The Reduced Enrichment for Research and Test Reactor (RERTR) program has successfully developed low enriched fuels for research reactors but does not address the large quantity of highly enriched fuels stored at locations around the world. In 1996, the U.S. initiated a program for the return of research reactor fuel with U.S. origin fuel material to the U.S. for interim storage and final disposition. The *Foreign Research Reactor Spent Nuclear Fuel Acceptance Program's* objective was to reduce the U.S. origin HEU stockpile in research reactors globally.

In parallel to the U.S. history and during the Soviet era, Russia similarly provided nuclear technology throughout the region and fabricated fuel for research reactors at Bochvar Institute in Moscow and the Chemical Concentrates plant in Novosibirsk. During the 1970's the Russian government became concerned with the quantity of HEU provided and used at research reactors throughout the Soviet Union and began to make changes by initiation of the *Russian Research Reactor Fuel Return Program.* Currently the U.S. and Russian Governments are cooperating to reduce and secure the HEU supplied inventory at research reactors under this program.

The U.S. Department of Energy (DOE) is sponsoring the development of a technology that could provide an alternate disposition path for Former Soviet Union (FSU) research reactor fuel if, for any reason, a research reactor's fuel cannot be shipped back to Russia as part of the *Russian Research Reactor Fuel Return Program*. Argonne National Laboratory (ANL) and Savannah River National laboratory (SRNL) have proposed the Mobile Melt-Dilute Technology to fulfill this mission need. This technology could provide an interim disposition path to mitigate proliferation concerns during the long logistical process that will be required to return the fuel to Russia from eighteen different countries.

The Mobile Melt-Dilute (MMD) process is an extension of the Melt-Dilute process that was developed at the Savannah River National Laboratory for treatment of spent reactor fuel of U.S. origin. Extension of the MD process to a mobile platform has resulted due to the many positive benefits of the technology which include the potential for significant volume reduction; reduced criticality potential, and proliferation resistance. By incorporating proven concepts, along with a closed system approach, a simplified mobile treatment system is being developed for rapid deployment. A summary flowsheet schematic is shown in Figure 1. The MMD process simply involves 1) loading spent fuel assemblies in a canister with depleted uranium, 2) welding a lid on the canister, 3) drying and evacuating the canister, and 4) melting the HEU fuel assemblies and diluting the ^{235}U/U alloy to less than 20%.

After treatment, the sealed canister containing the solidified aluminum-uranium ingot, can be placed in interim storage pending reprocessing or emplacement into long-term storage. Thus, HEU material can be treated using the MMD process to generate a safe and secure LEU ingot. As envisioned, the MMD system will be compact and staged on a transportable vehicle, with the capability to treat and encapsulate research reactor spent fuel at either the reactor or storage site.

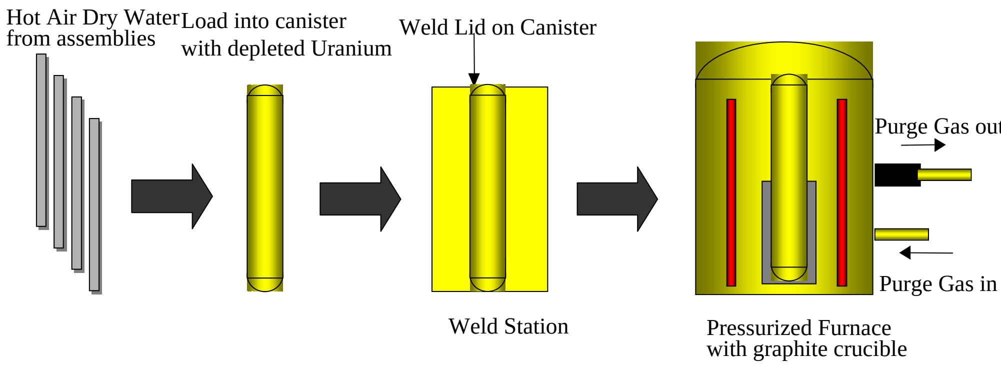

Figure 1. Schematic of simplified flowsheet for MMD process.

3. Process Layout

The MMD system consists of a treatment module and control module that are contained in International Standards Organization (ISO) transportable cargo shipping containers. The containers will be transported to the treatment site on two trailers. The system includes loading equipment and a transfer cask, welding equipment, vacuum systems, and an induction furnace. Electrical power is supplied by a diesel generator, and the induction furnace is air cooled eliminating the need for cooling water at the treatment site. Movement of the transfer cask is performed using a fork lift truck which will supply back up emergency electrical power. The treatment module will contain the induction furnace and support equipment such as the lid welder and canister evacuation system. The treatment processes will be operated remotely from a control module located inside an adjacent cargo container.

The furnace will be enclosed and radiation shielding installed to protect equipment and personnel. A bottom-loading shielded transfer cask is used to move fuel assemblies from the storage area in the reactor site to the MMD melt cell. The cask will attach to the furnace, fuel is then lowered into a canister in the furnace, and the bottom shield will be detached and secured to seal the furnace. The door can be closed to completely seal the canister inside the furnace.

After loading the canister with spent fuel, it is heated and dried under vacuum at 500°C to remove all adsorbed and chemically attached water from the aluminum base fuel assemblies. Some volatile fission products maybe released if heated near the fuel blister temperature or if failed fuel is treated. Water vapor and volatile fission gases released during drying will be trapped on special filters and will not be released to the atmosphere. After drying, the canister is sealed under vacuum prior to melting at 750 ºC. The sealed canister will contain the radioactive ingot and any gaseous or condensed volatile products inside after treatment. When cooled to ambient temperature, the canister will be moved to the final wet or dry storage facility.

The Direct Electric Heat (DEH) furnace, a patented technology of Inductotherm Corporation, was selected for the MMD process. It is a compact air cooled induction furnace that provides the capability to rapidly heat the fuel assembly and to stir the molten alloy to uniformly dilute the highly enriched uranium (HEU) fuel. The technology is ideal for the mobile process because of its small footprint and because minimum utilities are needed at the reactor fuel storage site. A sketch of the MMD system layout is shown in Figure 2.

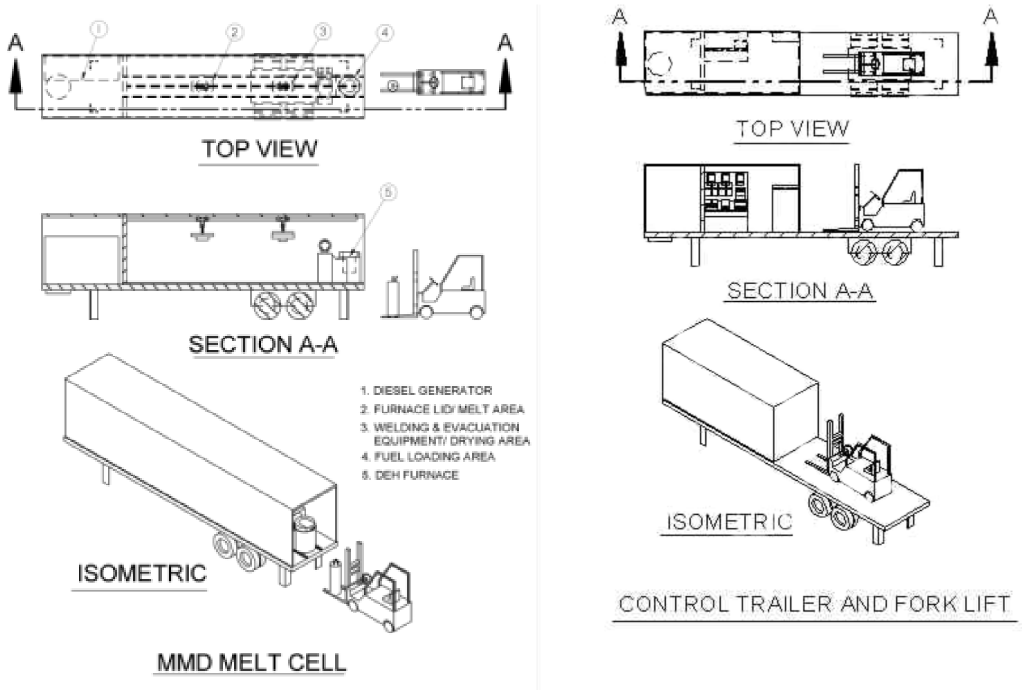

Figure 2. Sketch of MMD module layout.

Proof of principle experiments and component testing are currently underway to collect the necessary engineering data so that prototype designs of the furnace and canister can be prepared and fabricated for continued development of the MMD system. The following section describes the results of the key component and experimental test programs being performed to aid the design, fabrication, and demonstration of a prototype system.

4. Experimental Testing

Uranium Dissolution Tests

Uranium dissolution tests have been performed to determine preliminary processing characteristics. Dissolution tests were performed at 850, 800 and 750°C using commercial 6061 aluminum and a 4 wt% uranium in aluminum alloy, which is the expected composition of the melted spent research reactor fuel assemblies. Preliminary stirring tests were then performed to ensure that the required dissolution rate and uniform melt could be obtained at the lowest temperature of 750 °C.

The experimental tests were performed in a laboratory induction furnace using a carbon steel crucible. The furnace operated at 3000 Hz for melting and 60 Hz for induction stirring. A depleted uranium cylinder with high surface area was added to the molten alloy after reaching the test temperature. The dissolution characteristics were determined by taking samples from a fixed position near the bottom of the melt as a function of time for up to 30 minutes. The samples were analyzed using either Inductively Coupled Plasma Spectroscopy (ICPS) or X-Ray Florescence (XRF).

The dissolution rates for uranium in molten aluminum (4 wt% U and pure Al) without stirring are shown in Figure 3 and exhibit a power law decay with time. Diffusion of U in molten aluminum is a function of the melt temperature; increasing with increasing temperature.

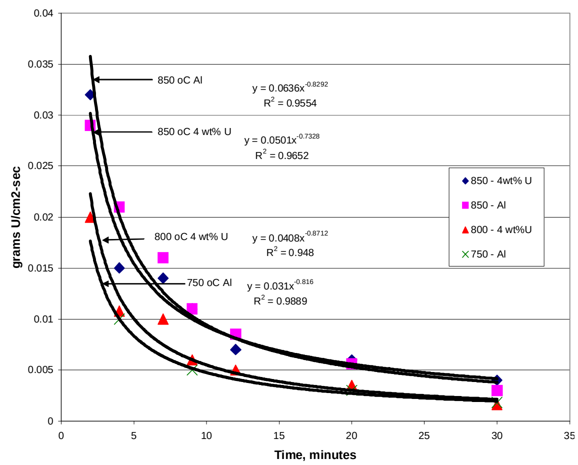

Figure 3. Dissolving Rate of Uranium in 6061 Aluminum and 4 wt% Uranium-Aluminum Alloy [4].

The sealed canister will meet Section VIII of the ASME Boiler and Pressure Vessel Code [3]. Below 815 ºC stainless steel can be used for pressure vessels according to the ASME pressure vessel code [3]. Testing has also shown that the dissolution rate of uranium in molten aluminum is also a function of the rate of mixing agitation and surface area of the diluent. In order to process at a lower temperature, stirring and optimized diluent feed surface area are needed. Initial tests performed at 750 ºC with melt stirring for 5 minutes show good uranium uniformity and higher diffusion rates. Tests at the lower operating temperature with prototypic components will continue to optimize stirring rates and feed characteristics to increase the initial diffusion rate.

5. Furnace Testing

A schematic of the preliminary canister and furnace design is shown in Figure 4. The canister design and process operating parameters require determining the DEH furnace heating characteristics at 750 ºC, the established melt temperature. The melting time should be about 1 hour at temperature to minimize molten aluminum and canister crucible reactions. The furnace is insulated so cooling rate of the canister is about 2 ºC/minute and depends on heat loss and air flow around the furnace coil. A melt cycle time of about 8 hours per melt is needed for the MMD process to meet throughput requirements.

Preliminary tests were performed using surrogate fuel assemblies made of aluminum. Four assemblies in a batch were placed in a stainless steel canister with a carbon steel canister liner containing surrogate fuel material was heated in the DEH furnace to determine heating characteristics. The surrogate assemblies were based on a Russian IRT type fuel assembly and were fabricated from aluminum plate and instrumented with 5 thermocouples equally spaced along its length on the outer and inner plates of the assembly. Temperature measurements were made by attaching thermocouples to the stainless steel canister wall at 25 cm and 56 cm from the bottom and by attaching thermocouples to the carbon steel crucible wall at 46 cm from the bottom of the crucible.

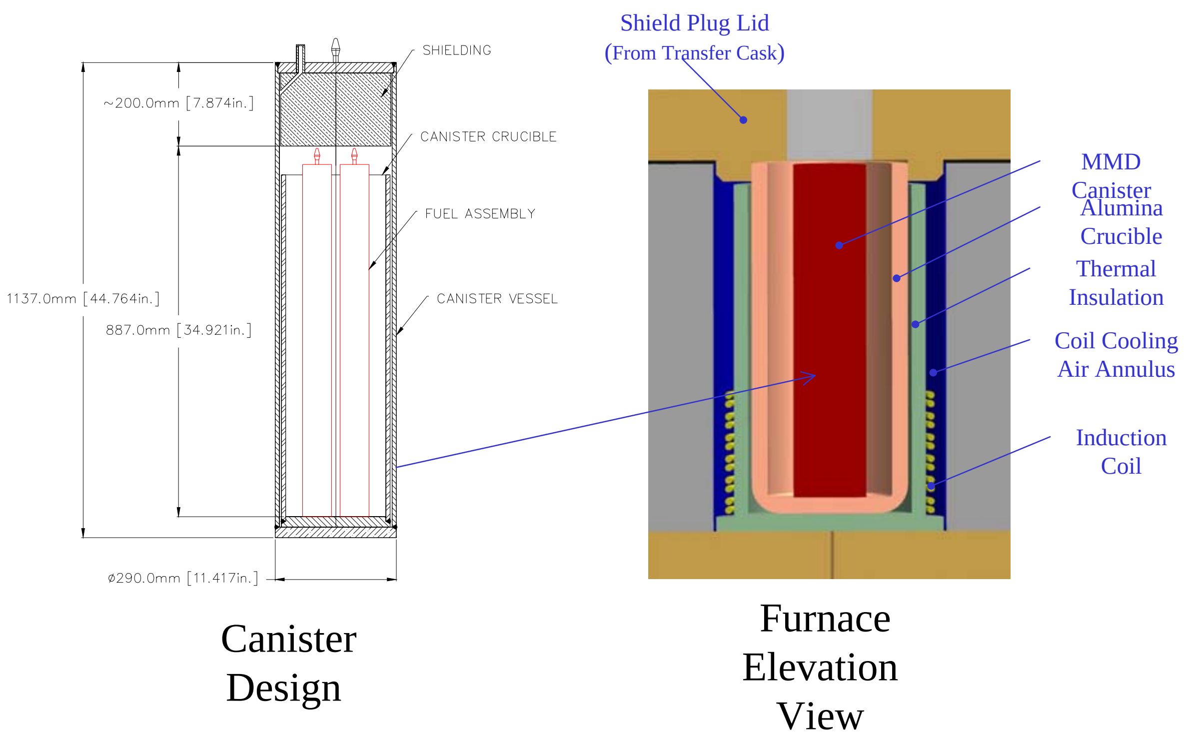

Figure 4. Schematic of preliminary design of canister and furnace.

The furnace power was originally 100 kw but after 5 minutes it was reduced to 40 kw. After the hot zone reached approximately 880 ºC, the peak temperature, the furnace power was reduced to 12 kw for the remainder of the test. The stainless steel canister maintained a temperature range of 740 to 800 ºC during processing that was well within the ASME pressure vessel code limit of 815 ºC. The temperature range for the carbon steel liner was 700 to 780 ºC. The heating cycle time was 120 minutes.

A temperature profile of the mock fuel assembly is shown in Figure 5 for the outer and inner plates. The bottom of the fuel assembly was in contact with the canister/crucible and heated faster due to conduction of heat. The top was heated slower from radiation and from heat conduction along the plates.

After 80 minutes the top of the fuel elements reached 500 ºC, the drying temperature. By this time the bottom of the fuel assembly had reached the melting point of aluminum as indicated by the isothermal line. Decreasing the initial furnace power gives a slower and even heating rate for the fuel assembly, which will reduce the potential for melting the aluminum cladding during the drying step. Note that the thermocouples at the top of the fuel assembly increased rapidly to 800 ºC after reaching the melting temperature. The thermocouples at the top pulled out as the fuel assembly began to slump and touched the side of the hot liner; therefore, the bottom thermocouples reflect the true melt temperature.

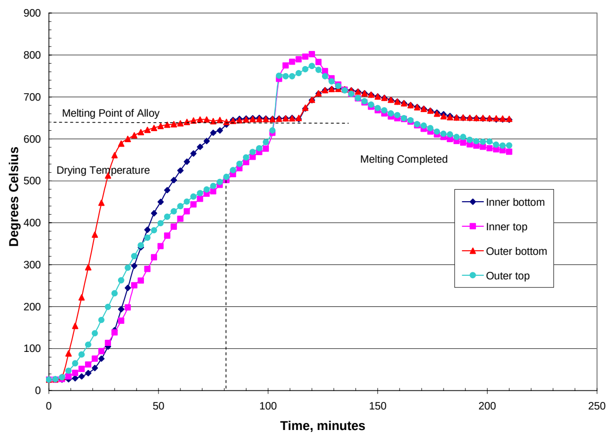

Figure 5. Temperature Profile for the Outer and Inner Plates for the Russian IRT Mockup Fuel Assembly during the DEH Test [5].

When the furnace power was shut off the molten aluminum began to cool. The cooling rate was calculated to be 2 ºC/minute and the cooling time was about 1 hour to the aluminum solidification temperature. The total cycle time from startup to complete melting was 3.5 hours during the test. Assuming the constant cooling rate, it would take approximately 9 hours to treat the spent fuel and to reach room temperature if cooled in the furnace.

6. Canister Liner Testing

A testing program was performed to evaluate candidate canister crucible materials to select a material that will withstand the high temperature environment and corrosive constituents of the Al-U molten alloy. The testing program is organized into two phases; short-term screening tests with several candidate crucible materials; and confirmatory testing with high potential candidates under simulated conditions.

For the first phase of canister crucible testing, two heating cycles were performed; the first at 850°C and the second at 750°C. Several small crucibles were loaded with approximately 100 g of aluminum (alloy 6061) and heated to the maximum temperature for 3 hours in an air atmosphere. Details about the crucibles used in these two tests are given in Table 1. Graphite was excluded from the matrix due to the air atmosphere and the potential for significant oxidation at the test temperatures. With the exception of the alumina castable crucible, the small crucibles were 2 inches in diameter by 2 inches in height. Nominal wall thicknesses varied for the crucibles according to the whether the original materials were milled or from tube stock. Floor thicknesses were initially 0.12 inches for the iron-based crucibles. The castable crucible was approximately 4 inches in diameter by 2 inches in height and was cast according to vendor specifications.

As noted in Table 1, two oxidized carbon steel crucibles were tested along with the others at 750°C and 850°C. The crucibles were oxidized or "blued" to potentially hinder the reaction of the crucible with the aluminum melt by forming a resistant oxide layer between the two. Approximately 0.1 g of weight gain was recorded for the two crucibles during the oxidation cycle. Crucibles 4-5 and 4-6 had additions of graphite and iron chips, respectively, to also inhibit the reaction of aluminum with the crucible surface. Iron additions greater than 1% have shown promise in minimizing the tendency of aluminum to solder during die-casting operations [7].

Table 1. Crucible Testing Materials and Conditions[6].

Table I. Crucible Matrix for Phase I Testing

Crucible ID	Material	Maximum Temp.(C)	Initial Weight (g)	Initial Wall (in)	Notes
1-1	304 SS	850	214.34	0.110	Welded, tubing and floor
1-2	304 SS	750	215.91	0.110	Welded, tubing and floor
2-1	316 SS	850	290.30	0.165	Welded, tubing and floor
3-1	410 SS	850	234.96	0.125	Milled
4-1	Carbon Steel	850	247.77	0.130	Milled, air oxidized for 4 hrs @ 500C
4-2	Carbon Steel	750	240.86	0.130	Milled, air oxidized for 17 hrs @ 500C
4-3	Carbon Steel	850	247.20	0.130	Milled
4-4	Carbon Steel	750	246.73	0.130	Milled
4-5	Carbon Steel	750	246.31	0.130	Milled, addition of 0.6 g graphite
4-6	Carbon Steel	750	244.16	0.130	Milled, addition of 5.1 g iron chips
None	Alumina Castable	850	558.25	0.700	Grefcon ® 98T, ~98% Alumina

Following the testing of smaller crucibles in Phase I, two larger crucibles were produced from carbon steel and tested either with a furnace located at Argonne or using the furnace expected for production operations. Larger crucibles were used to obtain a higher molten aluminum mass to exposed surface area ratio of 9-17 g/cm^2, which is comparable to full-scale operations. The test at Argonne was performed with uranium and aluminum (alloy 6061) in an induction furnace in an argon atmosphere. The other large crucible was tested with just aluminum (alloy 6061) at Inductotherm Corp. located in Rancocas, New Jersey, using an induction furnace system similar to the anticipated full-scale MMD operations furnace.

Results from the canister testing indicate that no significant loss of crucible material was measured for both the small and large carbon steel crucibles tests. As a result, carbon steel was selected as the material of choice for the MMD canister crucible based on its availability, ease of fabrication, and compatibility with aluminum and uranium at high temperatures.

7. Furnace Design

The MMD prototype furnace design is guided by furnace evaluation, information from the dissolution and integrated tests, and the Functional Design Requirements (FDR) for the MMD system. The prototype furnace will be used to precisely map power settings, time, temperature requirements, and stirring times to process the IRT surrogate fuel assemblies (the largest unaltered assemblies that fit the canister) and other fuel geometries in the MMD melt canister. The location of temperature controller transducers will be determined for the furnace and canister (if required for the canister). These transducers will allow the operator(s) to observe the temperature plateau of phase transitions as the fuel becomes liquid, and again as it freezes. A view of the MMD prototype furnace is seen in Figure 6.

The prototype furnace is specifically designed to process aluminum clad fuel assemblies and will be capable of processing multiple assemblies simultaneously. In the field, quantity will be determined by the fuel type and activity. Up to four IRT assemblies will fit simultaneously in the treatment canister.

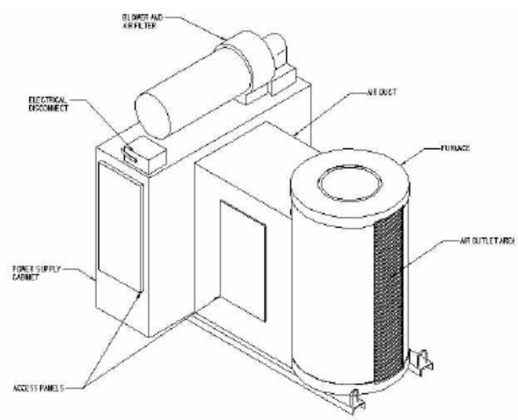

Figure 1: MMD Prototype Furnace

Figure 6. Prototype MMD Furnace Design[8].

Induction furnaces are normally rated by crucible and power supply capacity. Induction furnace power supplies fall within a range of operating frequencies designed to meet user needs. The furnace load geometry must also be considered so melting, pouring, ladling or other operations can be accomplished. Therefore, standard induction furnaces are tailored to the user's needs by adjusting both physical and electrical parameters. MMD requirements include specific canister geometry, efficient energy transfer to the canister, enough power to melt 100kg of aluminum quickly, and magnetic stirring. The MMD prototype will also be used to test air flow and power cable diversion caused by (mock) lead shielding. Inductotherm used their tools and experience along with input from the SRNL-ANL team and the results of the integrated test to match specific induction furnace parameters to MMD system needs. In list form, these are:

- Air-cooled induction furnace
- Furnace power supply output frequency 500 Hz
- Induction (magnetic) stirring circuit included in power supply
- Induction stirring frequency (pulse rate) •60 Hz
- Coil height: 28.5 inches
- Coil diameter: 20 inches
- Liner (crucible) inside diameter: 13 inches
- Cooling air flow: 2300 scfm
- Shunt coverage •80%
- Power supply rating 115 kW

- Duct length between power supply and furnace coil: 26 inches (minimum)
- Mounted on support frame (skid)
- Provided with 480V 3• 60Hz electrical disconnect attached on/near the power supply
- Remote furnace/temperature control capability

8. Summary

Currently, aluminum-based Russian research reactor fuel containing highly enriched uranium (HEU) is stored underwater in storage basins at various locations in FSU countries. The Mobile Melt-Dilute (MMD) treatment technology is an attractive option to convert the HEU weapons-useable fuel into a low enriched uranium (LEU), nonproliferation form, and encapsulate the fuel in a sealed container suitable for continued wet or dry storage and/or eventual reprocessing or disposal. The MMD process flowsheet was developed and comprehensive component testing and initial experimental test programs have been performed to guide the development of a prototype furnace and canister for the MMD system.

9. References

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